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Use of CFD for CO_2 absorbers optimum design : from local scale to large industrial scale

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Abstract

Post combustion CO_2 capture with chemical solvents is performed in packed columns operating in the counter current mode. Good predictions of the pressure drop and of the mass transfer characteristics is critical for their design. Many experimental works are dedicated to characterization of packings. However, experiments are carried out at a laboratory intermediate scale, with almost no possibility to bring information neither at small scale nor at very large scale. It is shown in this paper how CFD, via an original multi scale approach from the liquid film thickness to the industrial column dimensions, can help in complementing experimental data.

Keywords : CO_2 capture ; CFD ; packings ; VOF method ; wetting

1. Introduction

In the case of post combustion CO_2 capture with chemical solvents, flue gases are treated by liquid solvents in packed columns operating in a counter current mode, the gas going in the upward direction with the liquid flowing downward. In such an operating mode, a first design criterion concerns the diameter which has to be large enough to prevent flooding. Since the amount of flue gas to be treated would be so important that it requires very large size absorbers, high capacity packings are thus required. When using chemical solvents, such as amines like the monoethanolamine (MEA), the CO_2 undergoes an equilibrated reaction with the amines in the fast chemical regime. In this regime, mass transfer from gas to liquid is a limiting step that is the absorbed mass flux is limited by the effective interfacial area, a_e, between the gas and the liquid solvent and requires high mass transfer coefficients, k_G and k_L, in the gas phase and in the liquid phase respectively. In the precise case of CO_2 absorption with MEA, Tobiesen et al. [1] have shown that the mass transfer resistance is mostly located in the liquid side, the main parameter being the effective interfacial area. The second criterion is thus to develop a high effective area to minimize the height of the absorbers. High efficiency packings are then required. A power plant of 630 MWé may

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require up to 6 columns of 9 m in diameter and 30 m in total height. The cost of the equipped absorbers and desorbers represents about 40 to 50% of the total investments for the capture plant; the energetic cost due to the use of a booster fan to overcome the pressure in the process corresponds to 5 to 10% of the operational costs. It is thus of high importance both to predict local performances in terms of pressure drop and mass transfer parameters. It is also crucial to make sure that the design at large industrial scale is able to match the performance obtained with laboratory pilot tests performed at intermediate scale. The two requirements of low pressure drop and high efficiency are met by structured packings. Achieving full characterization of such packings is commonly done via intensive experimental work. Many works have been dedicated to pressure drop measurements and to mass transfer measurements and their modelization via 1D approach (see e.g. Billet [2]). In most cases, such models have been developed for air/water system or for light hydrocarbons systems characterized by low viscosity and low surface tension. Since the optimization of CO_2 post-combustion capture will require the use of new solvents, either with higher amine concentration or with original molecules having different physical properties, in particular high viscosities and moderate surface tension. Besides, it is known that the effective area in structured packing is less than the geometric area (Rocha et al. [3]) which calls for intensive studies on wetting phenomena. It is thus of high interest to study the influence of liquid physical properties on hydrodynamics and mass transfer in structured packings. It has been done for some packings (see e.g. Sidi-Boumedine and Raynal [4] or Tsai et al. [5]); however, testing the influence of each parameters (packing geometry and material, liquid physical properties, flow operating conditions...) requires intensive experiments in limited ranges and without being easily able to dissociate effects of each parameter from one another. This is a first major advantage of using CFD which makes possible complete sensitive studies for any parameter. The second major advantage of using CFD is to be able to address design issues at very large scale. Most experiments are actually dealing with column diameters ranging from 100 mm to 1 m while industrial absorber diameter would of the order of magnitude of 10 m. In experimental works, measurements are carried out with high quality distributing devices for further ensuring homogeneously distributed flows (see e.g. Alix and Raynal [6]), while this may not be the case at industrial scales.

In recent years, CFD has been more and more used for studying hydrodynamics in structured packings, however most of previous papers focus either on flow at small scale, either on flow at large scale without filling the gap between the different approaches. From CFD two-dimensional Volume Of Fluid (VOF) simulations at small scale, Raynal et al. [7] have shown that, in the case of very high liquid loads, liquid holdup could be significantly higher than the one deduced from the commonly used laminar film model. More recently, Ataki and Bart [8] made similar CFD simulations for an original structured packing, the Rombopak 4M but with a three-dimensional approach considering fully periodic elements. At a meso scale, Petre et al. [9] proposed a first approach based on calculations within said representative 3D elementary units, called REU. As discussed in Raynal et al. [7] such calculations open a nice route towards CFD modeling of flows in structured packings but they cannot be considered for quantitative purposes first because the considered REU geometry does not fully correspond to a representative geometry and second because the liquid is not taken into account, results thus apply to wet pressure drop only. Considering simulations at large scale, Wehrli et al. [10] have shown that CFD could be an appropriate tool for determining the gas flow field above the distributor and below the packed bed, but latter calculations did not take into account the packed bed itself. A strategy that makes a continuous link from small to large scale, that is that outputs obtained at a given scale are further used as inputs at a larger scale, has recently been proposed by Raynal and Royon-Lebeaud [11] for the particular case of the Mellapak 250.Y structured packing. This strategy involves calculations at three scales, small, meso an large scale, those scales referring respectively to the liquid film scale, the smallest periodic element in the packing and the column itself. Each scale requiring an appropriate type of modelization. The present paper follows the study of Raynal and Royon-Lebeaud [11] and shows complementary work dealing with simulations at small and large scales. Concerning simulations at small local scale, the present work not only complete the latter study for VOF 2D simulations but also discuss original results for VOF 3D simulations dealing with wetting phenomena. Concerning simulations at large scale, original gas distributor effects on the flow field within the layers of packings are discussed. These two steps, simulations at small scale dealing first with wetting phenomena and second with film flow characteristics and simulations at large scale are described in section 2. Sections 3 and 4 deal with the results obtained at small scale and at large scale respectively.

2. Calculation strategy

The gas liquid flow in a packed column is very complex and involves a large range of length scales. As shown in Figure 1, the gas/liquid flow at the industrial scale that is from inlets to outlets and in particular the gas distributor and the packed bed interactions are characterized by a length scale of several meters (see sketch a). Since each layer of packing are turned by each other with a 90° angle, an intermediate scale is the layer height of 21 cm. More importantly the characteristic size magnitude of the smallest periodic element as sketch in Figure 1.b (square with dotted lines) is a few centimeters; this corresponds to meso scale. Last when looking closely to the packing wall, that is at small scale or at liquid film local scale, packings may have perforations or texture, the size of which is close to a few millimeters as illustrated by the picture of Figure 1.c. For obvious CPU requirements reasons, it is presently impossible to solve all flow dynamics in such a complex geometry involving a length scale ratio of about 1:100,000 from the liquid film thickness to the column diameter and an adapted calculation strategy must be used.

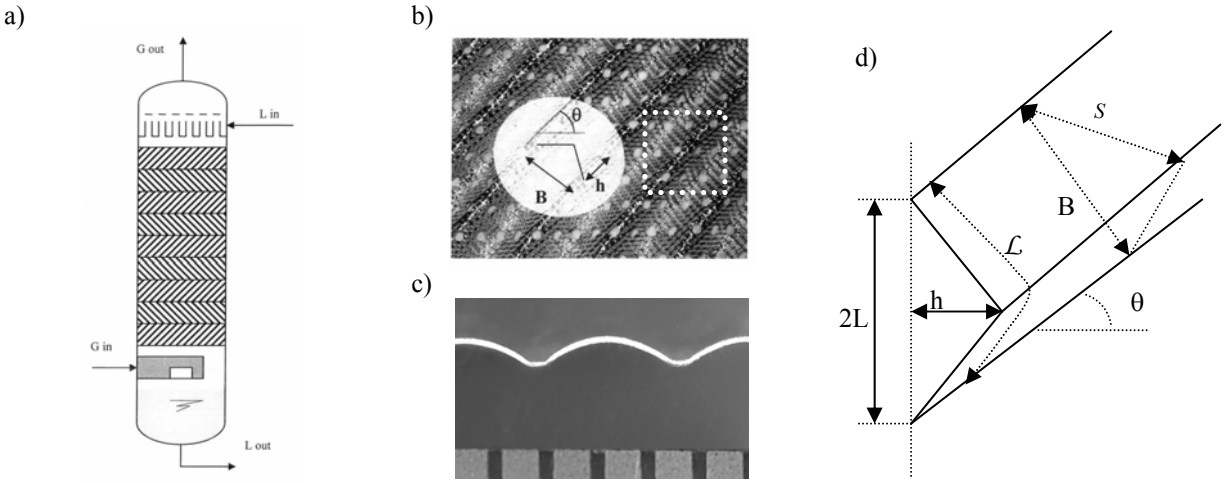

Figure 1 : Illustration of the multi-scale analysis; a) sketch of an absorber at large industrial scale with gas (G) and liquid (L) inlets and outlets, b) picture of the Mellapak 250.Y structured packing elements at meso scale, the square with dotted lines corresponds to the smallest periodic element of the packing, c) close picture of the wall texture of Mellapak 250.Y, the distance between two vertical lines being 1 mm, d) sketch of the packing at meso scale with notations used in the text.

Calculations at small scales consist in CFD calculations at the scale of the gas-liquid-solid interactions to pick up the effect of the surface wall texture and to catch the main characteristics of the liquid flow. The gas-liquid-wall interaction is simulated at the liquid film scale with the VOF approach. This two-phase flow model allows to capture the interface between two non-interpenetrating fluids. Simulations have been carried out with Fluent 6.2 commercial code. Two complementary studies are described : first a three-dimensional approach for better understanding of the wetting mechanism involved in structured packing, and second a bi-dimensional approach to characterize the liquid flow on a hydrodynamic and mass transfer point of view.

2.1. Calculation at small scale- wetting phenomena

The main purpose of the wetting phenomena study is the determination of the wetting ratio of the effective interfacial area a_e to the packing geometric area, a_G. Three dimensional simulations are compared to experimental results. Both numerical and experimental studies consider a liquid rivulet flowing over an inclined plate which material can be changed. A parametric study has been carried out by varying separately the liquid flow rate Q_L, the static contact angle θ, the surface tension σ, the injector geometry and the slope of the plate α.
The numerical domain is 31 mm in length, 24 mm in width and 3 mm in height. The liquid injection is of rectangular shape of size 2*1 mm^2 or 4*1 mm^2. An adaptative meshing method, based on the phase volume fraction gradient, is used with 15,000 to 45,000 initial cells depending on the injection size.

The experimental set-up consists in a planar solid plate with a slope angle with the horizontal, α wetted by water at a flow rate varying from 0.2 l/h to 60 l/h. Experimental changes in contact angle is obtained by changing materials. 2.5 or 4 mm diameter tubular and 20 mm width rectangular liquid injectors are used. A CCD camera combined with MATLAB image-processing methods yields the liquid envelop on the plate, from which one can measure the liquid rivulet width at different downstream positions.

2.2. Calculation at small scale- liquid film characterization

This study aims at describing the influence of the small-scale wall structure on the bi-dimensional liquid film flow. Indeed, and as shown in Figure 1.c, ones observes that the wall has texture on it, which can be represented as a sinusoidal-shaped wall, the periodic length and amplitude being respectively 2.8 and 0.3 mm for Mellapak 250.Y. The purpose of present simulations is to compare the liquid film characteristics, in terms of film thickness, e, and velocity at the interface, $U_{L,eff}$, to values given by the commonly used laminar film model. The latter model, corresponding to a uniform fully developed laminar film over a smooth vertical wall with no gas interaction, gives:

$$e = \left(\frac{3 q_L \nu_L}{g} \right)^{1/3} \text{ and } U_{L,eff} = \frac{3}{2} \left(\frac{g\, q_L^2}{3\, \nu_L} \right)^{1/3} \text{ with } q_L = \frac{Q_L}{a_G} \tag{1}$$

where q_L is the liquid flow rate per unit width of wetted surface, ν_L the liquid kinematic viscosity, g the acceleration of gravity, Q_L is the liquid load of the packing and a_G is the packing geometric area per unit of volume. For industrial conditions, the liquid load varies from approximately 10 to 100 $m^3m^{-2}h^{-1}$. This corresponds to liquid film thickness of a millimeter as a maximum. Since it is of same order of magnitude as the amplitude of the surface texture at the walls (A=0.3 mm), the latter has indeed to be considered. Since computational domain and numerical schemes are identical to those described in details in Raynal and Royon-Lebeaud [11], there are not given here.

2.3. Calculation at large scale

Calculations at large scale considers the packed bed as a porous bed which pressure drop characteristics are given by results obtained at meso-scale the latter using results obtained at small scale. Raynal and Royon-Lebeaud [11] have shown that changing the packing disposition could have an effect on the gas flow distribution at packed bed inlet for a given gas distributor. In the present case, the effect of the gas distributor has been looked at, the packed bed characteristics being unchanged. Since the methodology is identical to the one described by Raynal and Royon-Lebeaud [11], it is not described here.

3. Calculation at small scale - Results

3.1. Results at small-scale - wetting

Figure 2 summarizes the CFD parametrical study by volume fraction distribution on the plate. It shows the contours of the liquid rivulet for different operating conditions. One observes that an increase in liquid flow rate of 100%, between Figure 2 a and b, leads to an increase in wetted area of 70%. On the opposite an increase in contact angle θ from 24.5° to 67° between Figure 2 a and c leads to a reduced wetted area of 52%. Furthermore, varying the slope angle α, from 68.5 to 90° between Figure 2 a and e, increases the wetting by 13%. Whereas little effect is observed when the surface tension σ is multiplied by 2.2 (Figure 2 a and d) or when the injector width decreased from 4 mm to 2 mm (Figure 2 a and f). As a preliminary conclusion, the controlling parameters for the wetting mechanism are the liquid flow rate and the static contact angle.

Figure 2: Contour of the volume fraction at the plate surface for the different CFD simulations: a) Reference case Q=5.4 l/h, θ=24.5°, σ=29 mN/m, α=68.5°, L_{inj}=4 mm; all other cases have identical parameters values but the one given b) Q=10.8 l/h, c) θ=67°, d) σ=64mN/m, e) α=90°- f) L_{inj}=2 mm.

Experimental parametrical study in terms of contact angle and injection geometry is summarized on Figure 3. Experimental results confirm that a decrease in contact angle leads to an increase in wetting area as can be seen between Figure 3 a and b. Indeed the couple PMMA/water has a higher contact angle than the couple gauze-plate/water, meaning PMMA has a lower wettability. Even with large injection the liquid rivulet width on PMMA decreases rapidly to value lower than the width measured in the case of the couple gauze-plate/water. Figure 3 c and d point out the importance of the pre-wetting of stainless-steel which leads to a large increase in wetting area by reducing the contact angle which is not observed for PMMA. On the same figure, on observes, in good qualitative agreement with CFD calculations, that the liquid rivulet width increases rapidly close to injection and is almost constant at a short downstream distance. Typically, at 20 mm downstream the injection, there is almost no more increase in width; measurements of rivulet width as a function of operating conditions have thus been made at this distance. This width increase can range a ratio from 1 in the case of PMMA up to 20 in the case of stainless steel as compared to the injection width at inlet, L_{inj}. The rivulet width measurements are shown in Figure 4 versus the function, *f*, proposed by Ataki and Bart [9] and supported by the experimental work of Shi and Mersmann [12]. The latter *f* function depends on non-dimensional groups which are given by :

$$f(\mathrm{Re}_{inj}, We, Fr) = A \times Fr^{-0.044} \cdot We^{0.243} \cdot \mathrm{Re}_{inj}^{-0.026} \tag{2}$$

$$\text{with } \mathrm{Re}_{\mathrm{inj}} = \frac{\rho_{\mathrm{L}} \mathrm{U}_{\mathrm{L}} L_{inj}}{\mu_L}, \mathrm{We} = \frac{\rho_{\mathrm{L}} \mathrm{U}_{\mathrm{L}}^2 L_{inj}}{\sigma}, \mathrm{Fr} = \frac{\mathrm{U}_{\mathrm{L}}^2}{g L_{inj}} \tag{3}$$

the non-dimensional groups being respectively the Reynolds, the Weber and the Froude numbers at injection. It is observed from Figure 4 that for all pre-wetted surface and for CFD results this function seems to be adequate to describe the wetting measurements. Only the pre-factor *A* in Eq.2 needs to be changed accordingly to the material. This change in material corresponds to a change in contact angles for CFD calculations. Note that in the cases of non-pre-wetted walls, the liquid rivulet width is much less than the one measured for pre-wetted conditions, this is particularly true for stainless-steel. One also observes than when comparing stainless-steel and gauze material the wetting efficiency is inversed if walls are pre-wetted or not.
These measurements indicates that, when possible, experimental measurements or industrial operation should first be run at maximum liquid load before being set to nominal conditions to simulate as much as possible pre-wetted conditions.

Since a good agreement between calculations and experiments is observed in terms of rivulet width evolution, similar calculations should be performed in the future on more three-dimensional realistic geometries, more representative of structured packings. The purpose would be to determine the wetting efficiency that is the ratio a_e/a_G. This type of calculations is very demanding in terms of CPU resources and explains why the determination of

liquid film flow characteristics have been performed in a two-dimensional geometries as discussed in the next section.

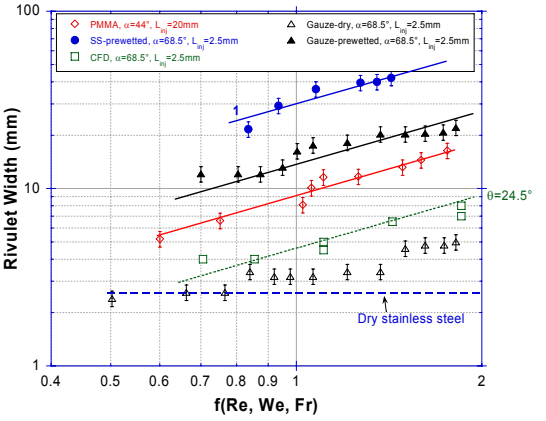

Figure 3: a) Front view of water rivulet and corresponding extracted envelop for a fixed liquid flow rate Q_L = 42.5m^3/h/m^2 and a constant slope angle α=68.5° with a tubular injector (L_{inj}=4mm) (except for a) PMMA α=44° and rectangular injection (L_{inj}=20mm) for different plate materials: a) PMMA ; b) prewetted gauze-plate, c) prewetted-stainless-steel, d) dry-stainless-steel.

Figure 4: CFD and experimental rivulet widths, at 20mm downstream the injector, as a function of f(Re_{inj}, We, Fr). PMMA: Q_L = 42→ 760m^3/h/m^2, Prewetted stainless steel: Q_L = 42→ 180 m^3/h/m^2, CFD: Q_L = 33→ 68 m^3/h/m^2, Gauze packing: Q_L = 5→ 180m^3/h/m^2

3.1.1. Results at small-scale - Characterization of the liquid flow

In this part the bi-dimensional flow over an inclined sinusoidal-like wall is considered. The calculation presented in Raynal and Royon-Lebeaud [11] are completed essentially to estimate the velocity at the interface for a large range of liquid flow rates and a large range of liquid viscosities. For this study with fixed amplitude of the wall texture and fixed Bond number, the relevant parameter is the film Reynolds number, defined as $Re_L=4q_L/\nu$.

Figure 5.a compares the liquid film thickness, e, determined at wall crest from two-dimensional CFD simulations to the thickness determined with the laminar film model (see Eq.1). It is observed that, for low Reynolds numbers, Re_L<200, CFD predicts a thickness slightly lower than the thickness evaluated by the one-dimensional laminar film model of around 0.9 times e_{1D}. As the Reynolds number increases the ratio of the CFD thickness to the 1D-model thickness increases and may reach up to 1.2 for Re_L>800.

Similarly, Figure 5.b compares the velocity at the interface obtained by both models. In the case of the CFD calculations, the velocity at the interface is determined by averaging local velocities over a period of wall oscillations. This averaging procedure is particularly needed for low Reynolds number for which the liquid interface follows the oscillations of the wall, which further implies periodic variations of the velocity as shown in Raynal and Royon-Lebeaud [11]. In the case of high Reynolds number, recirculation zones appears, and the velocity at the liquid film interface is not sensitive anymore to the wall texture and is almost constant. Figure 5.b shows the ratio of the velocity at the interface obtained by CFD normalized by the corresponding one as obtained by the laminar film model (see Eq.1). For Re_L<100, the mean interface velocity is slightly higher than the velocity predicted by the one-dimensional laminar film model and is close to 1.1. This ratio decreases as the Reynolds numbers increases down to 0.9 for Re_L>500.

These calculations show that the flow dynamics must be known in details in particularly for determining mass transfer characteristics. Indeed, the liquid velocity at the interface is further used for the liquid side mass transfer coefficient, k_L, determination (see Bravo et al.) via the Higbie theory using :

$$k_L = 2\sqrt{\frac{D_{CO_2,L}}{\pi\, t}} \quad \text{with} \quad t = \frac{\mathcal{L}}{U_{L,eff}} \tag{4}$$

where $D_{CO2,L}$ is the diffusion coefficient of CO_2 in the liquid phase and t is the exposure time. It is believed that exposure time can be written as the ratio of the renewal length, $\mathcal{L}$., to the liquid velocity at the interface, $U_{L,eff}$. The renewal length is believed to be the distance along the wall between two contact points as designated by Sidi-Boumedine and Raynal [4] and as shown in Figure 1.d. This distance is greater than the packing characteristic S as used by Rocha et al. [3]; it is given by the following relationship :

$$\mathcal{L} = 2\left(S^2 + \frac{B^2}{4}\tan^2\theta\right)^{1/2} \tag{5}$$

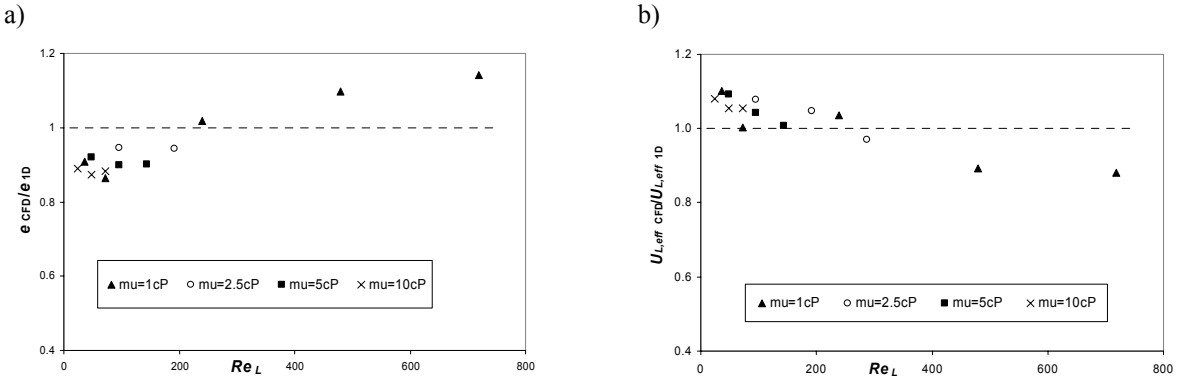

Figure 5: a), Ratio of the liquid thickness calculated by CFD at the top of the wall crest to the liquid thickness given by the one-dimensional model and b), Ratio of the mean liquid velocity at the interface calculated by CFD along a period of wall oscillations to the liquid velocity at the interface given by the one-dimensional model, both versus Reynolds number for 4 different liquid viscosities.

From these results, one can deduce that, in the case of low Reynolds numbers, using the laminar film model will lead to an under estimation of the liquid velocity at the interface which further will induce an under estimation of the liquid side mass transfer coefficient. This under estimation would be even more important that, for low Reynolds numbers, the wetting efficiency is low. One should indeed take partial wetting into account for modification of the liquid flow rate, it can be done via the relationship :

$$q_{L,pw} = q_L\left(\frac{a_e}{a_G}\right)^{-1} \tag{6}$$

where the subscript *pw* corresponds to partial wetting. On the contrary, for Reynolds numbers higher than 250, using the laminar film model will lead to an over estimation of the liquid side mass transfer.

4. Calculation at large scale - Results

Present calculations at large scale consider a packed column with a liquid load of 50 $m^3/m^2/h$, and a gas superficial velocity of 1.47 m/s. From calculations at small scale, one can compute the velocity at the interface being -0.5 m/s. This output of calculations at small scale is used as an input for calculations at meso-scale considering the real three-dimensional packing geometry; such calculations are not shown here but they are discussed into details in Raynal and Royon-Lebeaud [11]. From calculations at meso-scale, one deduce pressure drop correlations which are further used as inputs in calculations at large scale as pressure drop models for the packed bed considered as a porous zone.
Figure 6 shows velocity vectors in the symmetry plane of the column and more importantly the color map of the vertical component of the velocity vectors at bed inlet. Results have been obtained for three different gas distributors. It is observed that the pipe distributor gives bad results even when equipped with a baffle (case b and c respectively). On the contrary, it is the simplest geometry, that is the case with no distributor (case a), which gives best results.
Such calculations have been performed for a column diameter of 1 m. Since this step at large scale considers gas flow only, the liquid influence is indirectly taken into account via pressure drop laws in the porous zone

corresponding to the packing, one easily understands that similar calculations can been performed on a very large diameter columns, which makes optimum design possible.

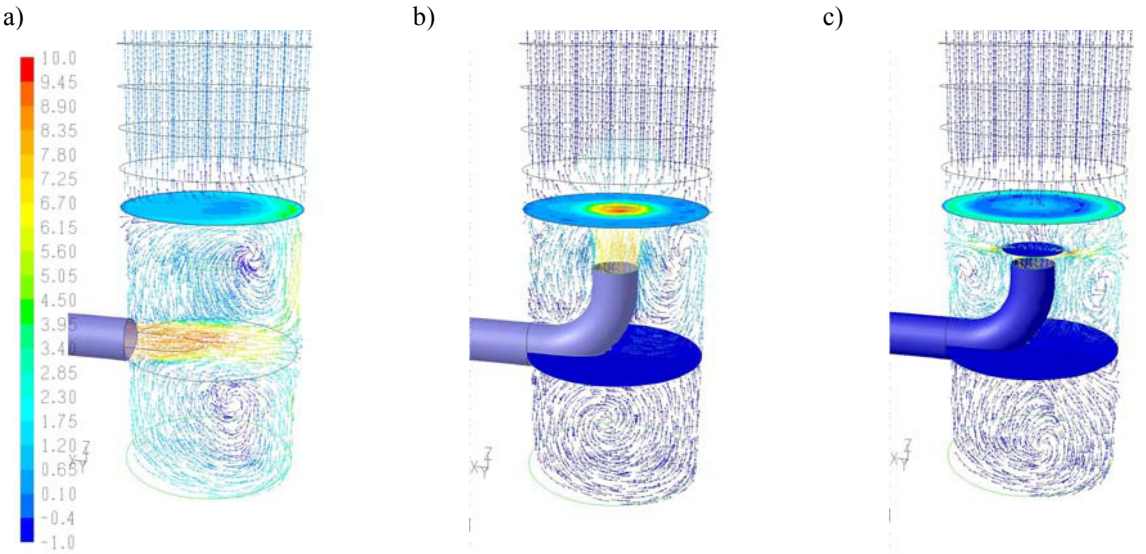

Figure 6 : Velocity vectors in the y=0 plane and vertical velocity contours at bed inlet. The colormap corresponds to the vertical velocity intensity. a) no distributor, b) vertical pipe distributor, c) vertical pipe with impact plate distributor. Q_L= 50 $m^3/m^2/h$ and F_S = 1.62 $Pa^{1/2}$.

5. Conclusions

It is shown in this paper how Computational Fluid Dynamics (CFD) can help in complementing experimental data. From CFD calculations at small scale, it is possible to test the influence of solvents physical properties on hydrodynamics and mass transfer which is very hard to do via experiments and may be of great help when developing original solvents as it is done in the EU Cesar project for example (www.co2cesar.eu). It also offers to tests various packing materials or packing geometries and eventually may help in the design of new ones. Using outputs from simulations at small scale, in particularly giving the wetted surface which further enables the determination of the velocity at the gas/liquid interface, as inputs in simulations at large scale, it is shown that CFD can be used for studying the interaction between the gas/liquid flow in the column and the distributors at industrial scale.

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